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BIODEGRADATION OF SILICATE AND ALUMINOSILICATE MINERALS: A LITERATURE REVIEW

by

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SUMMARY

A critical review of the literature on the biodegradation of silicates has been made. A number of microorganisms, of different species, has been reported to be capable of degrading minerals. The production of organic acid metabolites, such as citric, oxalic, 2-ketogluconic, tartaric, humic, fulvic and lichenic acids are involved, though not necessarily acting directly, in the biodegradation. Chelation is also one of the mechanisms postulated. Applicability of microorganisms for beneficiation of bauxite has been considered and areas for further work have been outlined.

BIODÉGRADATION DES MINÉRAUX CONTENANT DU SILICATE ET DE L'ALUMINOSILICATE: ÉTUDE DE LA DOCUMENTATION SCIENTIFIQUE

par

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RÉSUMÉ

Une étude approfondie de la documentation scientifique relative à la biodégradation des silicates a été effectuée. On signale qu'un certain nombre de micro-organismes appartenant à différentes espèces sont capables de décomposer les minéraux. La production de métabolites à partir d'acide organique tels les acides citrique, oxalique, 2-cétogluconique, tartrique, humique, fulvique et lichénique fait partie du processus bien qu'elle n'agisse pas directement sur la biodégradation. La chélation est également un des mécanismes susceptible de promouvoir la décomposition. La possibilité d'utiliser les micro-organismes pour valoriser la bauxite est à l'étude et d'autres secteurs d'application ont également été déterminés.

1. Introduction

The most abundant elements in the crust above the mantle of the earth (consisting of igneous rocks, sedimentary rocks and the hydrosphere) are oxygen (55.1%), silicon (16.3%) and hydrogen (15.4%) (1). The largest and most important group of minerals, constituting about 95% of the earth's crust, is the silicates (2). The majority of the silicate minerals occur in crystalline forms. In these minerals, Al^{3+}, Mg^{2+}, Fe^{2+} and Ca^{2+} can all be substituted for Si. The susceptibility to degradation of these minerals is controlled by the isomorphous substitution (2). The various aluminosilicates and associated minerals encountered in the survey and their formulae are listed in Table 1.

Biological activity plays an important role in the geological cycle for silicon. The main contribution of the biological activities in the Si cycle are indicated in Fig. 1.

Many aluminosilicates such as muscovite, biotite, leucite, orthoclase, microcline, sandstone and glauconite contain up to 13-20% potassium. Such aluminosilicates are so widely distributed that the mean content of potassium in the earth's crust reaches 2.58% (9).

It has been estimated that in a 1 metre layer of the soil, the amount of potassium in the form of aluminosilicates is of the order of 150-350 tonnes per hectare which is sufficient to meet the nutrient requirements of plants for thousands of years. As this potassium cannot be assimilated directly by higher plants, the study of the process of break-down of aluminosilicates in the soil is of enormous practical importance to agricultural and geological researchers. The former are interested in the break-down since a large supply of potassium fertilizer could be obtained therefrom and the latter are interested since it is a primary pedogenesis (soil-formation) process.

2. Microbiology

2.1 Bacteria: are unicellular organisms and, with a few exceptions, the bacterial cell is contained within a rigid or semi-rigid cell wall conferring a constancy of form. Cell multiplication involves growth and

division (asexual reproduction). In fluid environments many species are motile. Under conditions that are unfavourable for growth, some species of bacteria form spores (uni- or multi-cellular, asexual, reproductive or resting body that is resistant to unfavourable environmental conditions).

Organisms that need only inorganic compounds as nutrients for growth are called autotrophic organisms; organisms that require both inorganic and organic compounds are called heterotrophic.

2.2 Fungi: are plant-like organisms that lack chlorophyll (green photosynthetic pigment of plants) and are devoid of such organized plant structures as stems, roots and leaves. They include yeasts, mildews, molds and mushrooms. Fungi are also heterotrophic but their growth cycle is complex. Their vegetative body is formed of a great number of branching hyphae, tubular filaments that bear spores. The filaments, collectively known as mycelium, obtain food from organic matter.

2.3 Algae: (oxygen-evolving photosynthetic organisms in the plant subkingdom Thallobionta) a collection of pnyla, or divisions which exhibit a diversity of form, ranging from one-celled organisms as small as bacteria to elaborate giants such as the kelps, comparable in size with flowering plants. Diatoms, which belong to the class of algae, contain aluminum silicate in their walls.

2.4 Lichens: associations of fungi and algae, are among the most remarkable and successful examples of mutualism, a type of symbiosis, to be found in nature. They appear to be single plants, however, under a microscope, the association is seen to consist of millions of cells of algae woven into a matrix formed of the filaments of the fungus. Lichens are often confused with mosses which are plants of the class Bryatae, occurring in nearly all damp habitats except the ocean.

2.5 Bacteria, fungi, algae and lichens have all been implicated in the degradation of rocks and minerals, but most studies have been done on the organic-acid producing bacteria and fungi. Pedological studies (soil science), particularly the pioneer work in Europe and more recent work discussed later, have shown that many silicate minerals can be degraded by a

wide range of microbially produced organic acids. The microbes involved and the organic acids which they produce are presented in Table 2.

The mechanisms of aluminosilicate destruction by bacteria and fungi are connected with the action of different metabolites products of the chemical changes in living cells by energy is provided for the vital processes and activities of the cell secreted by the microorganisms. Groudev et al. (11) postulate that acidolysis (any chemical reaction, analogous to hydrolysis, in which an acid plays a role similar to that of water), chelation (producing organo-metal complexes, with a cyclic structure in which the central ion is a cation or a metal including Si) and solubilization by the alkali produced by microbes may be the mechanism involved (depending on the type of secreted metabolites).

Pioneering work in biodegradation of aluminosilicates was done by agricultural microbiologists. According to Rossi (25), DeGrazia and Camiola (5) were among the first investigators who studied biodegradation of aluminosilicates. However, Dacey et al. (2) imply that Muntz (6) in 1890 suggested that bacteria were involved in the degradation of rocks to form soil. Bassalik (7) in 1912 isolated from the intestines of earth worms bacteria (Bacillus extorquens), which had the ability to break down various aluminosilicates. Subsequently in 1913, he reported on an investigation in which 12 silicate minerals (orthoclase, microcline, oligoclase, labradorite, nepheline (elaeolite), leucite, muscovite, meroxene (biotite), olivine, augite, hornblende, tourmaline) and apatite, were biodegraded with several strains, including B. extorquens and yeast (8). The best results were obtained with strains of B. extorquens, with a dissolution of 78% in 146 days in the most favourable case, with 'Magnesia-mica'. The experiments were conducted with the minerals ground to -0.1 mm, and heated to about 300°C immediately before use.

Bacteria capable of biodegradation of silicate were isolated by Aleksandrov in 1939 directly from soils (9). Similarly, Novorossova et al. (10) isolated from soils bacteria which were capable of degrading aluminosilicates. According to Aleksandrov and Zak (9) and Novorossova et al. (10) Vernadskii (3) wrote repeatedly that the break-down of the aluminosilicates is due to the direct action of microorganisms on the chemical bonds rather than by the action of carbon dioxide and organic acids. Novorossova et al. used minerals ground to -0.25 mm and a medium containing sugar, aspargine

and ammonium phosphate. The maximum dissolution reported is 50% each of silica and alumina. These workers observed that the dissolution took place in about 10 days and that no further dissolution took place even though the experiment was extended to more than 30 days. In fact, they observed a decrease in the degradation. Aleksandrov and Zak (9) isolated two strains of bacteria, which were capable of breaking down aluminosilicates. One strain resembled the slime forming Bacillus mucilaginosus and the other resembled Bacillus megatherium, var. De Bary. Aleksandrov proposed the name "silikatnye bakterii" for the former. The so-called silicate bacteria were able to release over 75% of the potassium in the aluminosilicate in five days. These experiments were conducted in petri dishes. Media used for growth are given in the Appendix.

3. Microbial Action

3.1 Acid-producing Organisms

3.1.1 Bacteria

Only in a few cases has a positive identification been made of the microbe and the acid which it produces. Members of the genus Pseudomonas, particularly P. fluorescens, appear to be the important bacteria in producing 2-Ketogluconic acid. Bacteria of other genera have been reported to produce 2-Ketogluconic acid, however, the identification is not positive (12).

The name "silicate bacteria" has been used to indicate some bacterial isolates which were capable of biodegrading aluminosilicates (3,13). The bacteria are common soil organisms and all appear to be members of the taxonomic genus Bacillus. In more detailed recent studies it has been claimed that the so-called "silicate bacteria" are similar or even identical to bacteria previously described by other workers. Tesic and Todorovic (14) claim that most of the isolated bacteria are related to the species B. circulans described by Jordan in 1890. The bacteria B. circulans are typical soil microorganisms. Apparently, the different silicon-consuming isolates, including the "silicate bacteria" are a 'slimy phase' during the growth cycle of B. circulans or merely are its ecotypes and varieties (15,16,17). Groudev et al. (11) conclude that the name 'silicate bacteria'

does not have a taxonomic meaning and that the requirements of some varieties of B. circulans for silicon are so distinctly expressed (in comparison with those of other microorganisms) that this name is largely used to indicate those varieties and even the species B. circulans as a whole.

The silicate bacteria related to the species B. circulans, are rod shaped-cells 2.0 to 5.0 microns long and 0.7 to 1.5 microns wide; the shape and the appearance of their colonies grown on solid nutrient media depend on the medium composition (14,17). On synthetic nitrogen-free media containing silicates or aluminosilicates, the colonies observed are shiny glass-like buttons with one or more cells (larger than the normal ones) in a slimy capsule. Large amounts of Si and Al are accumulated in those capsules (15). The silicate bacteria are heterotrophic and need carbohydrates such as glucose and sucrose as substrates. Various organic acids mainly citric, gluconic and oxalic acids, which are formed from these carbohydrates during the metabolism of the bacteria are secreted into the culture media (18). The mechanism of aluminosilicate destruction by silicate bacteria is not fully understood but the capsule and acid formation play an important role (11,19).

3.1.2 Fungi

Fungi are well known organic acid producers - citric acid is produced industrially with Aspergillus niger. Numerous acid producing species of fungi have been isolated from weathered rocks (20,22). Zajic (23) found that 94% of calcium silicate and 96% of zinc silicate were solubilized by some fungi (unidentified). The fungus Penicillium simplicissimum, which produces citric acid, solubilized up to 80% of the titanium in granitic rocks but less than 2% of the titanium in basaltic rocks in the experiments carried out by Silverman and Munoz (24). The biogenic solubilization of titanium was not a function solely of the titanium content of the rock but also reflected the differences in susceptibility and the differences in the grain sizes of the titanium bearing rocks. Between 21 and 27% of the potassium contained in leucite ores from Central Italy was reported, by Rossi (25), to have been leached out in 150 days, with strains of Aspergillus niger, Penicillium expansum and Scopulariopsis brevicale. The leaching of these ores in sulphuric-acid-producing-thiobacilli, even in the presence of pyrite, did not give good results.

Kiel and Schwartz (26), leached a low grade copper ore containing chrysocolla ($CuSiO_3.2H_2O$) and malachite ($Cu_2[(OH)_2CO_3]$), with a copper content of 1.0-1.2%, with A. niger and observed 87.3% of the copper being solubilized in 2 weeks. Longer incubation time resulted in a decrease in the amount of copper in solution. Arieta and Grez (27) found that A. niger solubilized iron from augite, hornblende and biotite and postulated that complex formation could be the mechanism involved. Even such stable minerals as plagioclase and quartz were broken down by acid producing fungus P. notatum (28). The author emphasizes in his conclusion that the solubilization is not a result of direct action of the acid on the mineral. Karavaiko et al. (29) report that the fungi P. notatum and A. niger are capable of degrading spodumene, considered to be a highly stable mineral, with the liberation of lithium. Avakyan et al. (30), observed that regardless of the chemical composition of the rocks and minerals, fungi accelerated the extraction of Li, Si, Al and Fe in comparison to the purely chemical process. Strains of fungi, A. niger and P. simplicissimum whose main metabolic products were citric acid and citric and oxalic acids, leached aluminum out of a variety of clay minerals such as kaolinite, halloysite, illite, montmorillonite, vermiculite, serpentine, chrysotile, genthite, etc. (18). The susceptibility of thermally activated minerals to biodegradation was much greater than that of non-activated raw materials.

3.1.3 Algae and lichens

Pedological studies indicate that the lichens, mosses and algae are the first, and often the only, colonizers of rocks at high altitudes (31). The mechanism of degradation by algae and lichens is again through the production of acids. Humic, fulvic and lichenic acids, which like the organic acids produced by bacteria and fungi, are able to form chelates with metallic ions. However, they are much less effective than citric acid.

Werner (32) has conducted some interesting experiments on the regulation of metabolism in diatoms by silicate. Lichens and algae are of very little interest for the solubilization of silica and aluminosilicates in the beneficiation of bauxite and hence will not be considered any further.

3.2 Microbes That Produce Alkali

Sarcena lutea and S. ureae, which produce alkaline metabolic products, particularly ammoniacal compounds, are capable of solubilizing nepheline, plagioclase and quartz (2). Kutuzova (33) compared the action of the acid formers (P. notatum and Pseudomonas sp) and the alkali former (S. ureae) and found that the amount of Si dissolved from nepheline by Pseudomonas sp. was almost seven times that by S. ureae.

3.3 Organisms as a Sink for Weathering Products

Living organisms may also ennance weathering processes by acting as a sink for the soluble products of weathering. Lauwers and Heinen (34) reported that monomeric silica, resulting from the mineralysis of polymerized silica or quartz, was taken up by P. mirabilis cells and also by the higher plant Equisetum. Tesic and Todosovic (15) reported an absolute nutritional requirement for Si by silicate bacteria and the accumulation of Si in their slime layers. The ability of living cells to accumulate large concentrations of potassium against a concentration gradient is well documented. Eno and Reuszer (35,36) reported the accumulation of potassium in the mycellium of A. niger when grown in the presence of biotite, muscovite, greensand and microcline. Weed et al. (37) observed that fungi weathered biotite, muscovite and phlogopite to vermiculite by acting as a sink for the potassium released from these minerals.

3.4 Rate and Extent of Biological and Organic Chemical Weathering

The rates of biological and organic chemical breakdown of silicates are difficult to assess in the field, since it is not possible to differentiate between the effects of the two processes. Hence, most studies have been conducted in the laboratory. Most of the studies reported have investigated the action of a single biological species or of an organic chemical in an isolated environment. The concentration of one of the inorganic constituents of the rock or mineral is then taken as a measure of weathering. Goldich (38) formulated a weathering stability series (see Fig. 2), concerning the stability of minerals in a weathering environment. Table 3 lists the quantity of some of the mineral forming elements released to solution from these minerals by pure cultures of organisms. Comparison of the data in the table is difficult due to variations in the procedures.

The weathering of rocks and minerals in nature is a slow process, taking place over geologic time periods, whereas biological and chemical processes which break down silicates in the laboratory take place within days or weeks. Much more fundamental research needs to be done before an industrial process to beneficiate bauxite by the solubilization of silica biologically can be envisaged, since there is insufficient data for scale-up.

4. Possible Mechanisms of Silicate Degradation

Acidolysis and chelation are two important mechanisms by which microbes could influence the degradation of rocks and minerals.

4.1 Degradation by Organic Acids

Dacey et al. (2) cited the work of Mateeva who suggested that minerals can interact with aqueous solutions in two ways. Firstly, anions and cations from the mineral can become hydrated and pass into solution. Secondly, H^+/OH^- ions in the solution may attack the mineral, the favoured mechanism being H^+ attack on the O atom in an Si-O-Al bond, with redistribution of the proton's positive charge to the Si and Al atoms, which is then attacked by OH^-.

$$\text{Si-O-Al} + H^+ \rightarrow \text{Si-}\overset{\delta+}{\text{O}}\text{-}\overset{\delta+}{\text{Al}}\text{-H}$$

The higher the concentration of H^+, the greater the extent of the reaction. There are many reports of degradation of silicates and rocks associated with a lowering of pH of the medium by the organisms involved (20,24,27,28,29,30). Living organisms increase the hydrogen ion concentration by the production of organic and inorganic acids. A list of the organic acids, other than fulvic and humic, which were found in the literature is given in Table 4.

Huang and Keller (39) studied the dissolution of silicon, aluminum, iron and magnesium in acetic, aspartic, citric, salicylic, tartaric and

tannic acids. Duff and co-workers (12,20,40) have studied the effect of microbially produced citric, oxalic and 2-ketogluconic acids on silicates. The silicate bacteria studied by the Russian workers and others (7,8,9,13,16) have also been shown to produce acidic metabolites.

4.2 Chelation

A few organic compounds of biological origin can chelate or bind a variety of cations and metals and thus solubilize relatively insoluble inorganic compounds including silicates (41,44). The groups -OH, -COOH, -C=O, $-CH_3$, -COOR, $-NH_2$ and R-NH are the ones that are met most frequently in chelating compounds (2). Arrieta and Grez (27) showed that unidentified substances in acidic fungal culture fluids chelated the iron released biologically from iron-containing silicate minerals. Boyle et al. (45) found that the amount of Fe and Al solubilized from biotite was greater when the chelating ability of the biogenic acid was greater. Huang and Kiang (46) found citric acid to be most effective organic acid in solubilizing Al and Ca, presumably because of its better complexing ability. Huang and Keller (47) observed that the total weight of clay minerals dissolved by strongly complexing organic acids exceeds that dissolved by distilled water by factors 5 to 75. The factors for Si and Al were 2 to 35 and 3 to 500 respectively. The same authors (48) observed that the relative distribution of pH-dependent aluminum ion species in aqueous solution is markedly altered by aluminum salicylate complexes within the pH range 1.5 to 7.5.

Organic compounds are capable of removing metal already released into solution by hydrogen ion attack on silicate minerals. In addition, they are also able to remove metals, from silicate minerals, directly by forming complexes. Iskander and Syers (49) reported that the amounts of Fe, Al, Ca and Mg released from biotite, granite and basalt were more than what could be accounted for as due to hydrogen ions alone. Appreciable quantities of Fe and Al were removed from augite, epidote, biotite and granodiorite by sodium salicylate and sodium citrate in the pH range 6.8 to 7.7 (50).

4.3 Other Mechanisms

Another process involved in the degradation of minerals, is oxidation/reduction. For example T. ferrooxidans will oxidize Fe^{2+} to Fe^{3+} and obtain the energy of the electron transfer process for its metabolism. In biotite

degradation the oxidation of ferrous iron to ferric iron has been reported to occur (31). It is possible that microbes are involved in the oxidation.

Another mechanism is where the living organisms could act as a sink for the soluble products of weathering, thereby enhancing the weathering process. Such a process is understandable since the ultimate source of inorganic nutrients for micro and macro flora must be obtained from rocks and minerals of the earth. Lauwers and Heinen (34) observed that monomeric silica, released by mineralysis of polymerized silica or quartz, was taken up by the higher plant Equisetum and the bacterium Proteus mirabilis. The accumulation of silica in slimes and silica as an absolute requirement for nutrition was reported by Tesic and Todorovic (15). An excellent review of the organic geochemistry of silica was made by Siever and Scott (51). Eno and Reuszer (35,36) reported that the fungus Aspergillus niger accumulated potassium in its mycelium during growth in the presence of biotite, muscovite, greensand and microcline. The release of potassium from these minerals was attributed in the past to a shift in potassium equilibrium as a result of the A. niger removing K. Weed et al. (37) demonstrated that fungi weathered biotite, muscovite and phlogopite to vermiculite by acting as a sink for K released from these minerals.

5. Discussion

The literature review indicates that microorganisms play a significant role in the degradation of many silicate minerals and rocks. Most authors believe that the metabolites (mostly acids) produced by the microorganisms are responsible for the attack on silicate mineral structures. The vigour with which the metabolic products attack rocks and minerals is attributed to their ability to form chelates with calcium, iron, magnesium and to a lesser extent, with aluminum and silicon. The susceptibility of the minerals (and rocks) to chelating agents are dependent on:

(a) the chemical composition
(b) the extent of substitution of elements in various lattice sites
(c) the lattice stability
(d) permeability
(e) the mineral grain size
(f) porosity

(g) the existence of cracks, cleavage planes and the existence of other lines of weakness.

The last four factors, which are physical in character, are applicable more to rocks than to minerals. Since most of the studies reviewed were conducted with finely ground minerals, the chemical factors (a) to (c) dominate.

Dacey et al. (2) collated the silicate minerals in increasing order of degradability as follows:

Increasing Degradability

quartz

kaolinite

microcline, orthoclase, albite

plagioclase, feldspar

montmorillonite, muscovite, chlorite, hornblende, hypersthene, diopside, augite, epidote

biotite, phlogopite

leucite, natrolite, genthite, vermiculite, wollastonite

nepheline, olivine

Table 4 (from Dacey et al. (2)) shows the susceptibility of minerals to attack by organic acids. Oxalic, citric, tartaric, salicylic and 2-ketogluconic acids, all of which are good complexing agents, seem to be the strongest. Oxalic acid is most effective on Mg^{2+}, $Fe^{2+/3+}$ and Al^{3+}; citric and 2-ketogluconic acids on Ca^{2+} and Mg^{2+}; salicylic and tartaric acids on Ca^{2+}, Mg^{2+}, $Fe^{2+/3+}$, Al^{3+} and Si^{4+}.

Heinen (52,53) is of the opinion that microbes which metabolize silica to form organic silicon compounds require already solubilized silica and only very small quantities are taken up even under favourable laboratory conditions. Factors which influence Si uptake rate are aeration, cell concentration, silica concentration and pH. Heinen (53,54) using ^{32}P found that the uptake of Si was preceded by a three-hour adaptation period. Heinen is of the opinion that Si partially takes the function of PH. He found that a concentration of 4.26×10^{-3} moles of PO_4 suppresses uptake of

Si. Maximum uptake was obtained with solution containing 1.2 g/L of glucose.

Andreev and co-workers (54,55,56,57,58,59,60) used material ground to about 0.1 mm in size and obtained about 60% Si solubilization. These authors advocate the replacement of the medium every 3-5 days, and believe that the organic metabolites of the organisms are the active agents in dissolution of Si and Al.

Heydeman et al. (66) recently isolated organisms from bauxite and laterite samples from Australia, India and Africa. One of the isolates corresponded to B. circulans and the others were of the genus Bacillus. The content of Kaolinite and mica were decreased by 50% in 37 days.

Groudev and co-workers (4,11,18,21,61,62) have done extensive work with strains of B. circulans and other organic acid producing organisms. In Groudev's opinion silicate bacteria is a mucilaginous phase during the growth of B. circulans. They also contribute to the view that production of acid as a metabolite is the mechanism involved in the solubilization of silica. However, they stress the importance of other metabolites in addition to acids. Groudev strongly believes that B. circulans and other organisms can be used for the beneficiation of bauxite.

6. Conclusions

1. A large amount of literature exists on the interaction of micro-organisms or the metabolites of microbes with minerals. However, the reports are largely concerned with agricultural and pedological aspects. A wide range of minerals, with particular emphasis on silicates, are covered in the reports cited.

2. A wide range of organisms, including bacteria, fungi and lichens have been found to biodegrade silicates. Several mechanisms, including the action of organic acid metabolites, have been proposed.

3. Organic acid attack on minerals involve both H^+ ion attack and chelation.

4. The "silicate bacteria" referred to by the authors of Russian literature cited appears to be related to B. circulans. However, not all strains of B. circulans are able to biodegrade Si.

5. The information available in the literature is inadequate to embark on a large scale process for beneficiation of bauxitic ores.

6. A number of problems are foreseen in the practical application of these processes. In particular:

   (a) large amounts of cheap organic matter would be required to produce adequate amounts of metabolites;

   (b) sterilization of large quantities of material, to ensure that only the desired organisms are present, would be impractical and hence it may be difficult to ensure good yields of the desired metabolites;

   (c) disposal of large volumes of spent organic matter without detrimental effects to the environment.

7. Further studies to elucidate information on the following are required:

   (a) selection and identification of suitable organisms;
   (b) mechanism of the solubilization;
   (c) optimum conditions for solubilization; and
   (d) the interference of other metals such as iron.

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Table 1.

Mineral	Formula
Boehmite	$AlO(OH)$
Gibbsite	$Al(OH)_3$
Andalusite Sillimanite Kyanite	Al_2SiO_5
Staurolite	$(Fe^{2+}, Mg, Zn)_2Al_9(Si,Al)_4O_{22}(OH)_2$
Topaz	$Al_2SiO_4(OH,F)_2$
Almandine	$Fe_3Al_2(SiO_4)_3$
Pyrope	$Mg_3Al_2(SiO_4)_3$
Hemimorphite	$Al_4Si_2O_7(OH)_2 \cdot H_2O$
Hornblende	$NaCa_2(Mg,Fe,Al)_5(Si,Al)_8O_{22}(OH)_2$
Augite	$(Ca,Na)(Mg,Fe,Al,Ti)(Si,Al)_2O_6$
Kaolinite Halloysite	$Al_4Si_4O_{10}(OH)_8$
Pyrophyllite	$Al_2Si_4O_{10}(OH)_2$
Biotite	$K(Mg,Fe)_3(Al,Fe)Si_3O_{10}(OH,F)_2$
Lepidolite	$K(Li,Al)_3(Si,Al)_4O_{10}(F,OH)_2$
Glauconite	$(K,Na)(Fe,Al,Mg)_2(Si,Al)_4O_{10}(OH)_2$
Clinochlore	$(Mg,Fe)_5Al(Si_3Al)O_{10}(OH)_8$
Orthoclase	$KAlSi_3O_8$
Leucite	$KAlSi_2O_6$
Microcline	$KAlSi_3O_8$
Olivine	$(Mg,Fe,Mn,Ni)_2SiO_4$
Genthite	$TNiO_2 \cdot 2MgO \cdot 3SiO_2 6H_2O$

Montmorillonite	$XY_6Z_{12}O_{30}(OH)_6 \cdot nH_2O$ where X is Ca, Li, Na; Y is Al, Cr^{2+}, Fe^{2+} Fe^{3+}, Li, Mg Ni, Zn; Z is Al, Si
Chlorite	$X_5Y_4O_{10}(OH)_8$ where X is Al, Fe^{2+}_3 Fe^{3+}, Li, Mg, Mn, Ni Y is Al, B, Si, Fe^{+3}
Vermiculite	$(Mg, Fe^{2+}, Al)_3(Al, Si)_4O_{10}(OH)_2 \cdot 4H_2O$
Cordierite	$Mg_2Al_4Si_5O_{18}$
Spodumene	$LiAlSi_2O_6$
Illite	$(K, H_3O)(Al, Mg, Fe)_2(Al, Si)_4O_{10}[(OH)_2 \cdot H_2O]$
Serpentine	$(Mg, Fe^{2+}, Ni)_3Si_2O_5(OH)_4$
Chrysotile	$Mg_3Si_2O_5(OH)_4$
Epidote	$Ca_2(Al, Fe^{3+})_3(SiO_4)_3(OH)$
Muscovite	$KAl_2Si_3O_{10}(OH)_2$
Phlogopite	$KMg_3Si_3AlO_{10}(F, OH)_2$
Oligoclase	$(NaAlSi_3O_8)_x(CaAl_2O_8)_y$ where x and y are variables
Labradorite	$(NaAlSi_3O_8)_x(CaAl_2Si_2O_8)_y$ where x and y are variables
Nepheline	$KNa_3Si_4Al_4O_{16}$
Tourmaline	$(Ca, K, Na)(Al, Fe^{2+}, Fe^{3+}, Li, Mg, Mn^{2+})_3$ $(Al, Ca^{3+}, Fe^{3+}, V^{3+})_6(BO_3)_5Si_6O_{18}(OH, F)_4$
Apatite	$X_5(YO_4)_3Z$ where X is Ca, Sr or Mn; Y is P or As and Z is F^-, Cl^- or OH^-
Chrysocolla	$CuSiO_3 \cdot 2H_2O$
Malacnite	$Cu_2(OH)_2(CO_3)$
Plagioclase	$(NaAlSi_3O_8)_x(CaAl_2Si_2O_8)_y$ where x and y are variables

Table 2. Microbes reported to cause biodegradation and the metabolic products claimed to be responsible for their activity (reproduced from Dacey et al. (2))

Acid-Producing Microbes	Metabolic Product or Mechanism	Reference in this paper
Bacteria		
Pseudomonas fluorescens	2-ketogluconic acid	22
" aeruginosa		-
" striata		-
Proteus Mirabilis		34
Other Pseudomonas species	2-ketogluconic acid	12
Clostridium pasteurianum		-
Erwinia freundii	lactic acid	-
Achromobacter species		3
Bacillus mucilaginous, subsp. siliceus		3
" megatherium De Bary		-
" oligonitrophilus		-
" salivarius		-
" caldolyticus		34
" cereus	) formic acid	
" polymyxa	) acetic acid	61
" licheniformis	) butyric acid	
" circulans	) lactic acid	
Undefined bacterial species		
Actinomycetes		
Nocardia species	succinic acid	22
Other species		
Fungi		
Aspergillus niger	) oxalic acid) fumaric acid) citric acid) glycolic acid) gluconic acid) succinic acid	20
" flavus		
" awamori		
Other Aspergillus species		
Penicillium notatum		
" simplicissimum	citric acid	-
" sp.	citric acid	-
" expansion	gluconic acid	34
Schwanniomyces occidentalis		

Table 2. Microbes reported to cause biodegradation and the metabolic products claimed to be responsible for their activity (reproduced from Dacey et al. (2)) (continued)

Acid-Producing Microbes	Metabolic Product or Mechanism	Reference in this paper
Spicaria sp.	) oxalic acid) acetic acid	
Botrytis sp.	) formic acid	19&20
Cephalosporium sp.	citric acid	
Trichoderma sp.	citric acid	
Fusarium sp.	citric acid	
Hormodendrom sp.		
Mucor sp.		
Margarinomyces sp.		
Monilia sp.		
Popularia sp.		
Pullaria sp.		
Sporotriochum sp.		20
Undefined fungal species		
Algae		
Nostoc species	) humic and fulvic	
Undefined species	) acids	
Lichens		
Parmelia conspersa	fumarprotocetraric acid	
R. geographicum		
U. pustulata		
Stereocaulon vulcani	lichenic acids	49
Cladonia squamosa	squamatic acid	
Undefined species		
Alkali-Producing Bacteria		
Sarcina ureae	Alkali formation	22
" lutea	NH_3 formation	
Nitrifying Bacteria		
Nitrobacter winogradskyi	$NO_2 \rightarrow NO_3$	
Nitrosospira briensis	$NH_3 \rightarrow NO_2$	

Table 2. Microbes reported to cause biodegradation and the metabolic products claimed to be responsible for their activity (reproduced from Dacey et al. (2) (continued)

Acid-Producing Microbes	Metabolic Product or Mechanism	Reference in this paper
Silica Metabolising Bacteria		
Escherichia coli	SiO_2 uptake)	29
Bacillus subtilis	" ")	
Proteus mirabilis	SiO_2 accumulation)	54
Bacillus caldolyticus	" ")	
Undefined bacteria		
Fungi Capable of Metabolising K^+ from Mica		
Aspergillus fumigatus		
Zygorhynchus moelleri Vuill		
Penicillium janthinellum Biourge		37
Rhizoctonia species		
Chaetomium cochlioides Palliser		
Trichoderma species		

Table 3. Microbial solubilization of elements from silicate minerals

Mineral	Element Solubilized	Percent	Time (days)	Reference
Olivine	Mg	54.0	8	12
	Mg	52.0	7	20
	Fe&Mg	60.0	15	63
Augite	Mg	4.6	8	12
	Ca	10.7	8	12
	Fe	0.74	21	27
Hornblende	Mg	4.0	8	12
Biotite	K	9.45	5	64
	Si	3.0	5	64
		14.0	7	20
		3.0	7	20
		4.5	6	64
Clay	Si	8.0	20	65
K-Feldspar	K	1.0	8	12
Muscovite	K	7.1	5	64
	K	10.0	8	12
	K	26.8	18	63
	Al	2.8	5	64
	Al	7.6	8	12
	Al	4.0	7	20
	Si	2.39	5	64
	Si	9.0	7	20
Kaolinite	Al	53.3	37	66

Table 4. Generalized Susceptibility of Minerals to Attack by Organic Acids (Reproduced from Dacey et al. (2))

Acid / Mineral	Ac	* Ac + Fo + Ox	Asp	Sal	* 2-k.g.	Tart	* Ci + Ox	* Ox + Ci	* Ci	* Various (Ci,Hum, Ful,HCl)	Ful
Quartz								-		-/±	
Orthoclase					-			-			
Microcline	±		±	±		±	±	-		-/±	
Albite					-			-			
Plagioclase	++		++	++/+++	±	++/+++		-			
Natrolite		+++			+			++	++		
Chabazite					±						
Leucite		++						+++/++++	-		
Nepheline		+++			+			+++/++++	++++	++	++
Kaolinite								-		-/±	
Serpentine											
Genthite		+++			+			+++	+++		
Montmorillonite								-		+	+
Vermiculite								+++	-	+	+/++
Muscovite	++	-	++	++	+	++	±	±	-	+	+
Phlogopite		±			±			+	±		
Biotite					-		++	+		+	+
Chlorite											
Hornblende					±			-			
Hypersthene					±			-			
Diopside					±			-			
Augite	+/++		++	++	+	++		-			
Wollastonite					+++			±			
Epidote					-			-			
Olivine	+++		+++	++++	+++	++++		+++	++		
Datolite					+						

Key: - unaffected, ± negligible attack, + slight attack, ++ moderate attack, +++ strong attack ++++V. strong attack

Ac - acetic acid, Asp - aspartic acid, Ci - citric acid, Fo - formic acid, Ful - fulvic acid, Hum - humic acid, 2-k.g. - 2-ketogluconic acid, Ox - oxalic acid, Sal - salicylic acid, Tart - tartaric acid.
* denotes microbially-produced acid

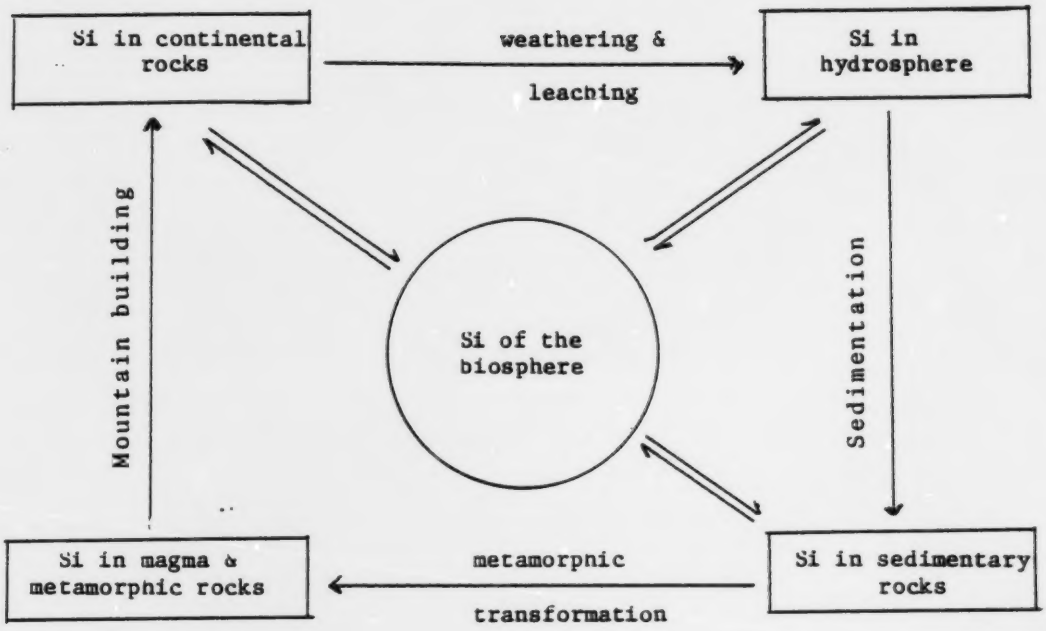

Fig. 1. Cycling of silicon in nature (from Ingri (1))

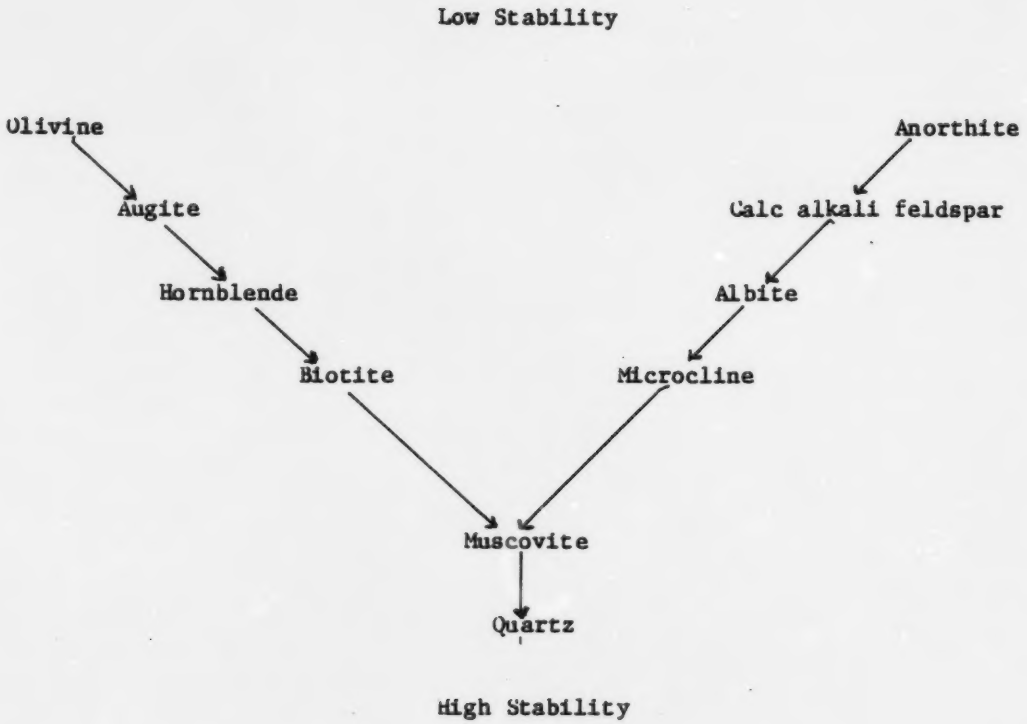

Fig. 2. A weathering stability series (Goldich (38))

APPENDIX

Media used for silicate bacteria

1. Mishustin et al. (67) No. 3

Ground glass	27.4%
Chalk	2.77%
Na_3PO_4	55.48%
$MgSO_4$	13.87%
$FeCl_3$	0.14%

4.2 g of this medium is in 1 litre of water

This medium was used by Aleksandrov & Ternovskaya (64).

2. Aleksandrov (13) No. 4

	Medium No.					
	1	2	3	4	5	27
Agar-aggar	2	2	2	2	2	2
Sucrose	0.5	0.5	0.5	0.5	0.5	0.5
$(NH_4)_2SO_4$	0.1	-	-	-	-	-
Na_2HPO_4	0.2	0.2	0.2	0.2	0.2	0.2
$MgSO_4$	0.05	0.05	0.05	-	-	0.05
$FeCl_3$	0.0005	0.0005	0.0005	0.0005	-	0.0005
$CaCO_3$	0.01	0.01	0.01	0.01	0.01	0.01
Potassium Alumina Silicate	0.1	0.1	-	0.1	0.1	-
Moscovite	-	-	0.1	-	-	-
Glass	-	-	-	-	-	0.1

Composition expressed as percentages

3. Aleksandrov et al. (64).

Aleksandrov's med. No. 27 with 100 mg of silicate was used.

4. Aleksandrov & Zák (9).

	Solid Medium	Liquid Medium
Sucrose	0.5	0.075
$(NH_4)_2SO_4$	0.1	0.015
Na_2HPO_4	0.2	0.03
$MgSO_4$	0.05	0.0075
$FeCl_3$	1 drop of 1% sol'n	trace
Aluminosilicate	0.1	0.1
Agar-agar (purified)	2.0	-

Composition expressed as percentages

5. Karavaiko et al. (29).

Aleksandrov's media T2 and A27 were used. See 2 and 8 (below)

6. Andreev & Kirikilitsa (58).

Medium A54 was used. See 13 (below).

7. Andreev et al. (57).

Nitrogen-free medium A27 was used.

8. Andreev et al. (54).

Aleksandrov's medium T-2 was used.

$MgSO_4$	6 g
NaCl	5 g
$FeCl_3$	1 drop of 1% sol'n
Na_2CO_3	400 mg
K_2CO_3	400 mg
Phosphorite flour with 10% chalk	2 g
Quartz sand with 10% chalk	8 g
Water	100 mL

9. Tesic & Todorovic (17).

Sucrose	0.5%
$(NH_4)_2SO_4$	0.1%
K_2HPO_4	0.2%
$MgSO_4$	0.05%
$FeCl_3$	traces
Aluminosilicates	0.1%
Agar	2%

10. Groudev & Genchev (21).

Medium described by Bogdanovic.

Distilled water	250 mL
Sand (< 100 microns)	20 g
Glycerol	5 mL
Agar-agar	5 g

11. Savostin (68).

	Solid Medium	Solid Medium	Liquid Medium
$Na_2HPO_4.7H_2O$	-	1.0 g	1.0 g
NaCl	-	0.2 g	0.2 g
NH_4Cl	0.5 g	-	
$CaHPO_4$	0.5 g	-	
$MgSO_4$	0.5 g	0.9 g	0.5 g
$FeCl_3$	0.09 g	-	0.09 g
$Ca(NO_3)_2.H_2O$	-	1.85 g	
CaCo	-	0.2	0.2 g
$(NH_4)_2SO_4$	-	-	1.0 g
Agar	17.0 g	17.0 g	-
Distilled water	1 L	1 L	1 L
pH	8.0	8.0	8.0

12. Karavaiko et al. (29).

Aleksandrov & Ternovskaya's
T-2 & A-27 media. See 2 and 8 (above)

13. Andreev et al. (60).

Medium A-54

Tap water	1 L
Apatite	20 g
Chalk	4 g
Quartz	20 g
Sucrose	5 g

14. Groudev & Genchev (61).

Liquid Medium

$NaNO_3$	2.0 g
KH_2PO_4	1.0 g
KCl	0.5 g
$MgSO_4.7H_2O$	0.5 g
$FeSO_4.7H_2O$	0.01 g
Sucrose	30.0 g
Distilled water	1 L
pH	7.0

Solid Medium

Sand (<100 microns)	20 g
Glycerine	5 mL
Agar	5 g
Distilled water	250 mL

15. Tesic and Todorovic (15).

Distilled water	1 L
Glycerine	20 mL
K_2NPO_4	0.2 g
$MgSO_4$	0.2 g
NaCl	0.2 g
K_2SO_4	0.1 g
$CaCO_3$	5.0 g
Agar	20.0 g

16. Andreev et al. (58).

Medium A.27

Agar	20 g
Sucrose	5 g
K_2HPO_4	2 g
$MgSO_4$	0.5 g
Potassium alumino silicate	10 g
Powdered quartz with 10% chalk	10 g
$FeCl_3$	10 drop of 1% sol'n
Water (distilled)	1 L

17. Heydeman et al. (66).

Nutrient Broth (NB)

Peptone	0.5%
Lab Lenco	0.4%
NaCl	0.5%
pH	7.0%

Nutrient yeast extract Broth (NYEB)

NB with the addition of 0.2% yeast

Glycerol Liquid Medium (GL)

Distilled water	250 mL
Glycerol	5 mL
Sand	10 g
Colloidal silica	1.2 g

Clycerol-ammonium liquid medium (GAL)

GL with the additin of 0.05% $(Na_4)_2SO_4$

Solid media were proposed by adding agar to a concentration of
1% in NA
2% in GL and GAL